The quantum Heisenberg antiferromagnet on the square lattice

Alessandro Cuccoli, Valerio Tognetti and Paola Verrucchi
Dipartimento di Fisica dell'Università di Firenze and Istituto Nazionale di Fisica della Materia (INFM), Largo E. Fermi 2, I-50125 Firenze, Italy

Ruggero Vaia
Istituto di Elettronica Quantistica del Consiglio Nazionale delle Ricerche, via Panciatichi 56/30, I-50127 Firenze, Italy, and Istituto Nazionale di Fisica della Materia (INFM)

The pure-quantum self-consistent harmonic approximation, a semiclassical method based on the path-integral formulation of quantum statistical mechanics, is applied to the study of the thermodynamic behaviour of the quantum Heisenberg antiferromagnet on the square lattice (QHAF). Results for various properties are obtained for different values of the spin and successfully compared with experimental data.

We consider the quantum Heisenberg antiferromagnet on the square lattice (QHAF), whose Hamiltonian reads

$$\hat{\mathcal{H}} = J \sum_{<\mathbf{ij}>} \hat{\boldsymbol{S}}_{\mathbf{i}} \cdot \hat{\boldsymbol{S}}_{\mathbf{j}} \ ; \tag{1}$$

J is positive, the sum runs over all the couples $<\mathbf{ij}>$ of neighbouring sites on the square lattice, and the quantum operators $\hat{\boldsymbol{S}}_{\mathbf{i}}$ obey the angular momentum commutation relations $[S^{\alpha}_{\mathbf{i}}, S^{\beta}_{\mathbf{j}}] = iS^{\gamma}_{\mathbf{i}}\delta_{\mathbf{ij}}\varepsilon^{\alpha\beta\gamma}$ with $|\hat{\boldsymbol{S}}_{\mathbf{i}}|^2 = S(S+1)$.

Several real compounds are well described, as far as their magnetic behaviour is concerned, by this model with $S = 1/2$ (La_2CuO_4, $Sr_2CuO_2Cl_2$), $S = 1$ (La_2NiO_4, K_2NiF_4) and $S = 5/2$ ($KFeF_4$, Rb_2MnF_4), and a consequently rich experimental analysis of the subject has been developed in the last ten years. From the theoretical point of view, an equally rich reservoir of results, from both analytical and numerical approaches, is now available; nevertheless, there are still many open questions, and different conclusions have been recently drawn by several authors [1,2,3].

To study the QHAF, we have used the pure-quantum self-consistent harmonic approximation (PQSCHA) [4], which is a semiclassical method based on the path-integral formulation of quantum statistical mechanics. Its main feature is that of exactly describing the classical behaviour and fully take into account the linear part of the quantum contribution to the thermodynamics of the system, so that the self-consistent harmonic approximation is only used to handle the pure-quantum nonlinear contribution. The fundamental goal of the PQSCHA is that of reducing the evaluation of quantum statistical averages to classical-like expressions involving properly renormalized functions, the fundamental one being the effective Hamiltonian $\mathcal{H}_{\rm eff}$. If $\beta = T^{-1}$, N is the number of lattice sites, $\boldsymbol{s}_{\mathbf{i}}$ is a classical vector on the unitary sphere ($|\boldsymbol{s}_{\mathbf{i}}| = 1$), and $\int d^N\boldsymbol{s}$ indicates the phase-space integral for a classical magnetic system, the quantum statistical average of a physical observable described by the quantum operator $\hat{O}$ turns out to be $\langle\hat{\mathcal{O}}\rangle = 1/\mathcal{Z}\int d^N\boldsymbol{s}\ \widetilde{\mathcal{O}}\ \exp(-\beta\mathcal{H}_{\rm eff})$, where $\mathcal{Z}=\int d^N\boldsymbol{s}\ \exp(-\beta\mathcal{H}_{\rm eff})$ is the partition function. Both $\mathcal{H}_{\rm eff}$ and $\widetilde{\mathcal{O}}$ depend on T and S, and the determination of their explicit form, starting from the expression of the original quantum operators, is indeed the core of the method [5]. The effective Hamiltonian for the QHAF, i.e. relative to Eq.(1), is

$$\frac{\mathcal{H}_{\rm eff}}{J\widetilde{S}^2} = \theta^4 \sum_{<\mathbf{ij}>} \boldsymbol{s}_{\mathbf{i}}\cdot\boldsymbol{s}_{\mathbf{j}} + \mathcal{G}(t) \ , \tag{2}$$

where $\widetilde{S} = S+1/2$, $t = T/J\widetilde{S}^2$ and $\mathcal{G}(t)$ is a uniform term that does not affect the evaluation of statistical averages. The renormalization coefficient $\theta^2 = \theta^2(t,S) < 1$ is easily evaluated, for any given t and S, by self-consistently solving two coupled equations [5]. ¿From Eq.(2) we see that the quantum effects leave the simmetry of the Hamiltonian unchanged and introduce an energy scaling factor θ^4, naturally defining the effective classical temperature

$$t_{\rm eff} = \frac{t}{\theta^4(t,S)} \tag{3}$$

that will enter all the PQSCHA results. The partition function, for instance, is $\mathcal{Z} = \exp[-\beta\mathcal{G}(t)]\mathcal{Z}_{\rm cl}(t_{\rm eff})$, where $\mathcal{Z}_{\rm cl}(t_{\rm eff})$ is the partition function of the classical model at a temperature $t_{\rm eff}$; $O_{\rm cl}(t_{\rm eff})$ will hereafter mean the value taken by the quantity O in the classical Heisenberg antiferromagnet at a temperature $t_{\rm eff}$.

The internal energy per site is easily found to be $u(t) = \theta^4(t,S)u_{\rm cl}(t_{\rm eff})$, while the correlation functions $G(\mathbf{r}) \equiv \langle\hat{\boldsymbol{S}}_{\mathbf{i}}\cdot\hat{\boldsymbol{S}}_{\mathbf{i}+\mathbf{r}}\rangle$, with $\mathbf{i} \equiv (i_1, i_2)$ and $\mathbf{r} \equiv (r_1, r_2)$ any vector on the square lattice, turn out to be

$$G(\mathbf{r},t) = \widetilde{S}^2\theta^4_{\mathbf{r}}G_{\rm cl}(\mathbf{r}, t_{\rm eff}) \ ; \tag{4}$$

the renormalization coefficients $\theta^2_{\mathbf{r}} = \theta^2_{\mathbf{r}}(t,S)$ are such that $\theta^2_{\mathbf{r}}$ does not depend on $\mathbf{r}$ for large $|\mathbf{r}|$, and $\theta^2_{\mathbf{r}} = \theta^2$ for $|\mathbf{r}| = 1$. From Eq.(4), we find the PQSCHA expression for the staggered susceptibility $\chi \equiv \sum_{\mathbf{r}}(-)^{r_1+r_2}\ G(\mathbf{r},t)/3$ to be

$$\chi = \frac{1}{3}\left[S(S+1) + \widetilde{S}^2\sum_{\mathbf{r}\neq 0}(-)^{r_1+r_2}\ \theta^4_{\mathbf{r}}G_{\rm cl}(\mathbf{r}, t_{\rm eff})\right] \ . \tag{5}$$

The PQSCHA result for the correlation length, defined by the asymptotic expression $G(\mathbf{r}) \propto \exp(-|\mathbf{r}|/\xi)$ for large $|\mathbf{r}|$, is $\xi(t) = \xi_{\rm cl}(t_{\rm eff})$, meaning that the correlation length of the QHAF at a temperature t equals that of its classical counterpart at a temperature $t_{\rm eff}$.

Once the problem has been reduced, by the PQSCHA, to a renormalized classical one, the ingredients needed to

obtain the temperature and spin dependent thermodynamic properties of the QHAF are the renormalization coefficients $\theta^2_{\mathbf{r}}(t,S)$, whose evaluation is a simple matter of a fraction of second on a standard PC, and the temperature dependence of the corresponding properties of the classical model, typically obtained by classical Monte Carlo simulations [6].

In the following we will focus our attention on the staggered susceptibility and the correlation length, as experimental data for these quantities are available for various compounds. Such compounds are usually characterized by a crystal structure in which the magnetic ions form parallel planes and mainly interact if belonging to the same plane; a weak interplanar interaction is responsible for a low-temperature 3D transition, and it introduces also an anisotropy term. Keimer et al. [7] have shown that in the classical limit, and to one-loop level, the relation between ξ in presence of the anisotropy term, and ξ_0 of the fully isotropic model, is given by $\xi = \xi_0/(1-\alpha\xi_0^2)^{1/2}$, where α is a parameter describing the relative strength of anisotropy; following Lee et al. [3] we shall employ the above formula (with some refinements [8] which lead to substitute α with its renormalized counterpart $\alpha_{\rm eff}$) to compare our PQSCHA results with experimental data.

In Figs. 1 and 2 we present our results for the correlation length of the QHAF for $S = 5/2$, and for the same quantity and the staggered susceptibility for $S = 1$. For $S = 5/2$ the experimental data refer to Rb_2MnF_4 [3] and $KFeF_4$ [9]: the anisotropy term is seen to be very well described by the approach described above. For $S = 1$ QMC [10] and experimental data for the two compounds La_2NiO_4 [11] and K_2NiF_4 [12] are reported.

We may thus confidently conclude that the thermodynamic behaviour of the QHAF is properly described by the PQSCHA, i.e. in term of a renormalized classical Heisenberg antiferromagnet. The easy-axis anisotropy, sometimes crucial to analyse the low-temperature experimental data, has been considered following Keimer et al. [7], in the PQSCHA framework. Finally, we would like to recall that our results help to shed some light on the reasons of the failure of the theory based on the nonlinear σ model approach [13], in describing the QHAF for $S \geq 1$ in the temperature region where experimental data are available.

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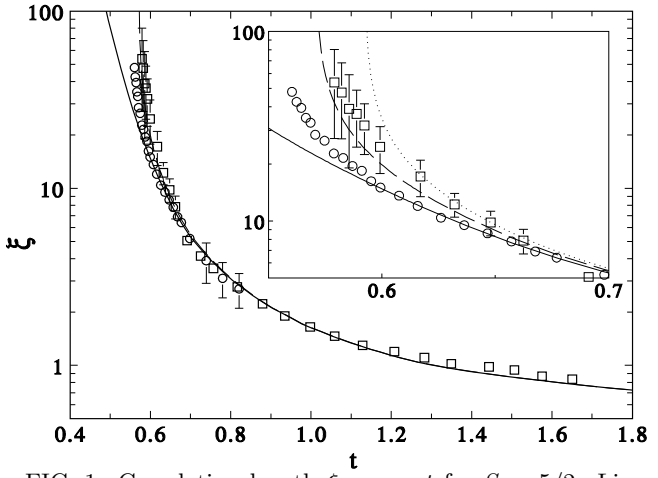

FIG. 1. Correlation length ξ versus t for $S = 5/2$: Lines are the PQSCHA results for the QHAF with the bare α (dotted), the renormalized $\alpha_{\rm eff}$ (dashed) and without anisotropy (full). Experimental data for Rb_2MnF_4 [3] (squares) and $KFeF_4$ [9](circles).

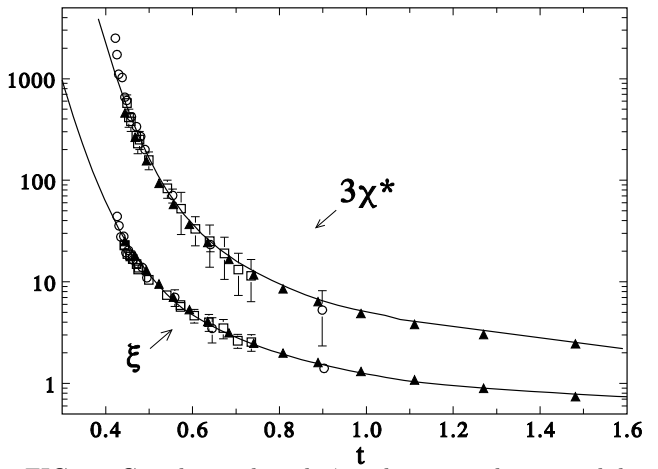

FIG. 2. Correlation length ξ and staggered susceptibility $3\chi^* = 3\chi/\widetilde{S}$ versus t for $S = 1$: Experimental data for La_2NiO_4 [11](squares), and K_2NiF_4 [12](circles); QMC data (triangles) from Ref. [10].